ANGULAR DEPENDENCE OF g-SHIFTS OF KRAMER'S DOUBLETS IN A CRYSTAL UNDER UNIAXIAL STRESS

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We measured the angular dependence of the g-shifts of the Γ_7 doublet in $Dy^{3+}:CaF_2$ under uniaxial stress. The data are compared with the predictions of a model for the orbit-lattice interaction.

When a stress is applied on a crystal doped with magnetic ions, the EPR lines from these impurities are shifted; measurements of these shifts can be used to evaluate the orbit lattice interaction. The strength of this interaction is given by the orbit-lattice coefficients, whose values are useful to predict spin lattice relaxation times and also contribute to the understanding of the natures of the interaction between the ions and the lattice [1].

The energy levels of a rare earth Kramer's ion in a cubic position transform like Γ_6, Γ_7 and Γ_8 in O_h. The Γ_6 and Γ_7 doublets show an isotropic g-value; when the crystal is deformed an anisotropic g-shift is produced by a second order perturbation process involving the orbit-lattice (H_{ol}) and Zeeman (H_z) interactions. This second order g-shift for a Kramer's doublet Γ_6 or Γ_7 is given by:

$$\Delta g = \frac{1}{\beta H}\sum_{i,s}\frac{2}{E-E_i}\,\mathrm{Re}\left(\langle\psi_{i,s}|H_z|\psi_a\rangle\langle\psi_a|H_{ol}|\psi_{i,s}\rangle + -\langle\psi_{i,s}|H_z|\psi_b\rangle\langle\psi_b|H_{ol}|\psi_{i,s}\rangle\right) \quad (1)$$

where ψ_a and ψ_b are the wavefunctions of the doublet and $\psi_{i,s}$ are the wavefunctions of the $\Gamma_8^{(i)}$ quartet, $s = 1,2,3,4$. H is the applied magnetic field, β the Bohr magneton, $E - E_i$ the difference in energy between the doublet and the i-quartet and Re means the real part of. Eq. (1) was obtained considering the transformation properties of $H_{ol}^2(\Gamma_1+\Gamma_3+\Gamma_5)$ and $H_z(\Gamma_4)$ in O_h, which gives non zero contributions only from the mixing of the doublet with the quartets. The wavefunctions of the doublet should be properly chosen to remove its Kramers degeneracy with H. The new wavefunctions depend on the direction of H given by the polar angles θ and φ, and thus Δg in eq. (1) is a function of these angles for a given H_{ol}.

We measured the g-shift of the Γ_7 doublet [3, 4] of $Dy^{3+}:CaF_2$ as a function of a stress P applied along the [110] direction with the magnetic field in the [110] plane; our data were obtained at 9.2 GHz and 4°K, and it is shown in fig. 1. Recently Baker and Currell [5] reported data for the same system, their value for $P/\!/[110]$ and $H/\!/[1\bar{1}1]$ is also shown in fig. 1.

The angular dependence of g obtained from eq. (1) for a Γ_7 doublet in our experimental situation is

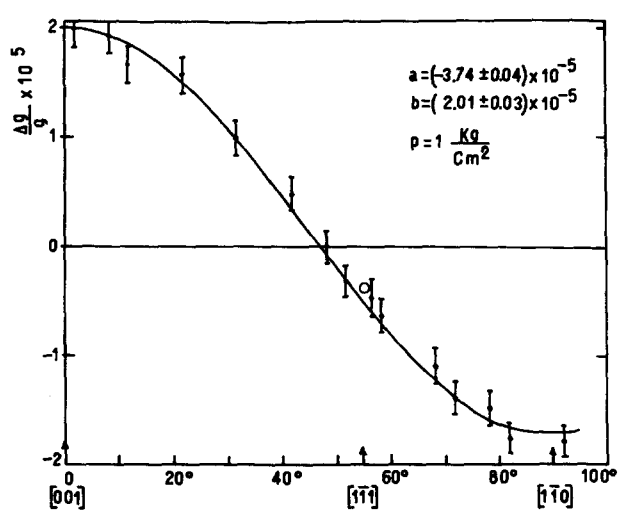

Fig. 1. Angular dependence of the g-shift of Dy^{3+} in CaF_2 under a stress P along the [110] direction. A value obtained in ref. 5 is indicated by a circle.

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$$\Delta g/g = a \sin^2\theta + b \qquad (2)$$

where θ is the angle between H and the [001] axis and a and b depend on the cubic field splitting and the orbit lattice interaction.

The measurement of $\Delta g/g$ as a function of θ provides a precise method for its evaluation; a least square fit of our data with eq. (2) gives

$$a = (-3.74 \pm 0.04)\times 10^5 \quad \text{and}$$
$$b = (2.01 \pm 0.03)\times 10^{-5} \text{ for 1 kg/cm}^2.$$

From symmetry considerations H_{ol} can be written as [2]:

$$H_{ol} = \sum_{n,i,\alpha,\xi} G_i^{(n,\xi)} O_{i,\alpha}^{(n,\xi)} \epsilon_{i,\alpha} \qquad (3)$$

where $\epsilon_{i,\alpha}$ is the linear combination of the components ϵ_{lm} of the strain tensor transforming like the α-component of the i-irreducible representation of O_h. The $O_{i,\alpha}^{(n,\xi)}$ is a linear combination of n-order Stevens' operators transforming accordingly and the $G_i^{(n,\xi)}$ are the orbit-lattice coefficients, ξ stand for the case where more than one Stevens' operator transform like $\Gamma_{i,\alpha}$.

Using eq. (1), (3) and (2) and the data on the cubic field splitting of the ion [3, 4, 6] we find for a and b the expressions:

$$a + \tfrac{3}{2}b = (193.74\, G_{5g}^{(2)} - 17730\, G_{5g}^{(4)} + \\ - 278200\, G_{5g}^{(6,1)} - 353040\, G_{5g}^{(6,2)})\times 10^8 \qquad (4a)$$

$$b = (-27.43\, G_{3g}^{(2)} + 14760\, G_{3g}^{(4)} + 309170\, G_{3g}^{(6)})\times 10^8 \qquad (4b)$$

where up to sixth order terms in H_{ol} are considered. Completely symmetric deformations contribute to the g-shift of the doublet mixing different Γ_7 doublets, a negligible contribution in this case.

To evaluate the $G_i^{(n,\xi)}$ and a and b from eq. (4a) and (4b) we used the ionic model of Sroubek et al. [1]; we find $a = -5.6\times 10^{-5}$ and $b = 3.6\times 10^{-5}$ considering the covalent correction [1]. This is in agreement in sign and order of magnitude with the experiments. Without the covalent correction we find $a = 10\times 10^{-5}$ and $b = 2.8\times 10^{-5}$ with a in disagreement both in sign and order of magnitude.

We conclude that the covalent correction is very important to fit our experimental data and improvements of the model taking this effect into consideration are needed. This agrees with Baker and Currell's conclusions [5] on the importance of the 4th and 6th order terms in H_{ol}, because these terms are the most sensitive to covalent bonding [7-9].

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